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# Interim Storage of Excess Plutonium: An Assessment of Options (U)

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# 1. INTRODUCTION

Recent agreements with Former Soviet Union (FSU) states will result in the removal of approximately 17,000 nuclear weapons from the active stockpile.[1] This development will dramatically alter the nuclear materials economy. In past decades, the weapons complex has been driven by frequent nuclear material shortages. However, the future will be characterized by an excess of plutonium. As such, storage of plutonium from these weapons has become one of the most important challenges for the nuclear weapons establishment. The nuclear materials effort must be redirected from one with emphasis on production and reprocessing of plutonium for stockpile needs to one with emphasis on maintenance of the enduring stockpile and on dismantlement of weapons and management of excess plutonium.[2]

Interim storage of excess plutonium is complex and includes both technical and political issues. Three general options have been identified for storage of excess material: (1) storage as intact pits, (2) storage as altered pits, and (3) storage in a suitable material form after extraction from the pit.[3,4] An adequate evaluation of these options must consider technical capability as well as facility requirements and availability. Differences in pit design (e.g., an outer container of stainless steel for certain units and beryllium outer shells for others) must be considered. Generation of radioactive waste is a particularly important component of environmental, health, and safety (ES&H) concerns related to these storage options. Additional issues with strong technical and political implications include rearmament and proliferation concerns. As a result, materials control and accountability (MC&A), transparency, and safeguards and security must also be addressed.

Readiness requirements and the time frame encompassed by "interim storage" are important factors for evaluating technical options. Both durations are somewhat uncertain. Implementation of various technical options within a period as short as 2 years has been initially suggested, while various time frames from 5 to 15 years have been subsequently proposed.[5] Technical options must also be examined in light of an interim storage period that will likely span several decades and could extend to as much as 100 years.

The focus of this report is a comprehensive technical evaluation of potential storage methods identified in a complex-wide assessment of storage options sponsored by the Department of Energy Arms Control and Nonproliferation office (AN).[6] As participants in that study, we encountered a wide diversity of opinion regarding the merits and suitability of various storage alternatives. As the coordinators of the DOE-AN study found, a comprehensive and internally-consistent report is difficult to prepare from input provided by several contributors with diverse perspectives. A credible report on plutonium storage must consider all pertinent information on physical and chemical properties of materials, the maturity levels of candidate technologies, potential hazards and complications associated with storage options, requirements for certification and packaging, and surveillance and monitoring of stored material. A comprehensive assessment must include relevant information from earlier studies by qualified experts as well as all potential options from diverse sources. Conflicting perspectives must be addressed. For example, non-proliferation advantages cited for certain chemically altered storage forms are inconsistent with a consensus of technical opinion stating that those materials can be used as nuclear explosives and that they are easily reconstituted to plutonium metal. Since our analysis focuses on technical issues, discussions of such topics as the process capability of specific facilities, material control and accountability (MC&A) issues, safeguards and security, and transparency concerns are limited. We attempt to identify and evaluate a range of technical issues associated with various options proposed for interim storage. Such an evaluation of storage forms cannot be divorced from the procedures and conditions required to obtain and certify those forms or from the procedures and conditions required to reconstitute the plutonium into a form suitable for fabricating a nuclear device.

## 2. DESCRIPTION OF STORAGE OPTIONS

### 2.1 Storage as Intact Pits

Storage of intact pits is an approach that employs the intrinsic characteristics that made the pit suitable for extended stockpile storage in a nuclear weapon. For an "intact" configuration, tubes might be coiled and immobilized to prevent torquing motion which could break welds or joints.[7] Although the title of this storage category seems to preclude any alteration of the pit, the possibility of removing tubulation has been suggested as a procedure to facilitate packaging. Additional storage options appear in the packaging configuration. Issues include the number and specifications for barriers between the pit and the ambient environment, and whether pit containers are stored within insulated shipping containers.

In contrast to metal or alloys, relatively large fractions of finely-divided forms are in the dispersible range.[103] 20–25 mass% of the $PuO_2$ formed by oxidation of the hydride consists of particles with dimensions less than 10 µm,[104] while 100 mass% of the oxide formed by pyrolysis of precipitates (e.g., hydrated plutonium oxalate) from aqueous processing is in this dispersible range.[105]

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Upon release of plutonium metal to the ambient environment, the potential for oxidation and subsequent dispersal by aerosolization exists. The oxidation rate of Pu in air at 500°C and above is modest and constant (0.2 g $PuO_2/cm^2 \bullet$ min of metal surface).[107] The oxide product formed at these temperatures is relatively coarse with less than 0.1 mass% in the dispersible range.[108]

Although container corrosion may also occur during a fire involving stored hydride, the possibility of container rupture exists for other powdered forms because of increased gas evolution and pressure and decreased container strength at high temperatures. The pressure is a combined effect arising from chemical reaction, radiolytic decomposition, helium release, thermal desorption, and thermal expansion. Rupture is expected to result in the dispersal of a larger fraction of powdered forms than would be released from massive metal or alloys.

On the basis of relative particle size distributions, the quantity of plutonium-containing material released to the environment is 1,000 to 10,000 times greater for finely-divided material forms than for massive metal. The risk of corrosion and container failure are likely reduced for alloy storage, but the potential hazard posed by such storage can not be assessed because information on reaction rates, thermodynamics, and particle distributions are not available.

Remediative actions can be employed to reduce the risks associated with containment of metal and powdered forms during a fire. Application of a ceramic coatings on internal container surfaces (e.g., erbium oxide)[109] or insertion of reactive container liners that alloy and chemically immobilize plutonium as a high-melting compound (e.g., aluminum)[110]

[89] G. Friedlander, J. W. Kennedy, and J. M. Miller, *Nuclear Radiochemistry, 2nd ed.,* John Wiley and Sons, Inc., New York (U) (1966).

[90] R. N. R. Mulford and B. A. Mueller, "Measurements of Helium Release from Materials Containing $^{238}PuO_2$," Los Alamos National Laboratory report LA-5215 (U) (July 1973).

[91] B. A. Mueller, D. D. Rohr, and R. N. R. Mulford, "Helium Release and Microstructural Changes in $^{238}PuO_2$," Los Alamos Scientific Laboratory report LA-5524 (April 1974).

[92] M. F. Stevens, T. G. Zocco, and J. H. Steele, "Bubble-Void Formation in Delta Phase Plutonium (U)," Los Alamos National Laboratory document MST-5-88-9 (SRD) (August 25, 1988).

[93] J. C. Martz, "Analysis of Plutonium Storage Pressure Rise," Los Alamos National Laboratory document NMT-5:92-328 (U) (July 1992).

[94] J. L. Stakebake, D. T. Larson, and J. M. Haschke, *J. Alloys Comp.,* **202** (1993) 251, LA-UR-93-0040 (1993), ibid.

[95] H. H. Van Tuyl, "Packaging of Plutonium for Storage or Shipment," (U) 1981, ibid.

[96] J. M. Haschke and J. C. Martz, "Metal-Oxide Chemistry and Storage," presentation to the DOE workshop on plutonium storage, Los Alamos National Laboratory controlled publication LA-CP-93-159, Albuquerque, New Mexico (U) (May 26, 1993).

[97] J. Lowe, private communication, Atomic Weapons Establishment, Aldermaston, UK (U) (December 17, 1992).

[98] J. M. Haschke and J. C. Martz, "Metal-Oxide Chemistry and Storage," (U) (1993), ibid.

[99] J. M. Haschke, "Evaluation of Source-Term Data for Plutonium Aerosolization," (U) (1992), ibid.

[100] J. L. Stakebake and L. M. Steward, *J. Electro. Soc.* **119** (U) (1972) 730.

[101] J. C. Martz, J. M. Haschke, and J. L. Stakebake, "Plutonium Pyrophoricity," Los Alamos National Laboratory document LA-UR-93-2655, manuscript submitted to *J. Nucl. Mat.* (U) (1993).

[102] J. L. Stakebake, *J. Less. Common Met.* **123** (U) (1986) 185.

[103] J. L. Stakebake and M. R. Dringman, "Absorption of Oxygen on Sintered Plutonium Dioxide," *J. Nucl. Mat.* **23** (U) (1967) 349.

[104] J. M. Haschke, "Evaluation of Source-Term Data for Plutonium Aerosolization," (U) (1992), ibid.

[105] J. D. Moseley and R. O. Wing, "Properties of Plutonium Dioxide," (U) (1965), ibid.

[106] D. Soderquist, J. Haschke, and J. Martz, "Fire Containment of Plutonium Metal," TPP Proposal submitted to DOE Complex 21 Reconfiguration Office, Los Alamos, New Mexico (U) (March 24, 1993).

[107] J. Mishima, "Plutonium Release Studies II. Release from the Ignited Bulk Metallic Pieces," Battelle Pacific Northwest Laboratory report BNWL-357, Richland, WA (U) (November 1967).

[108] J. Mishima, "Plutonium Release Studies I. Release from the Ignited Metal," Battelle Pacific Northwest Laboratory Report, Richland, WA, BNWL-205 (December 1965).

[109] A. G. Philips and J. M. Haschke, "Use of Ceramic Coatings for Corrosion Protection," Rockwell International report CD88-1910 , Golden, Colorado (U) (April 1988).

[110] F. H. Ellinger, et al., "Constitution of Plutonium Alloys," (U) (1968), ibid.

[111] H. H. Van Tuyl, "Packaging of Plutonium for Storage or Shipment," (U) 1981, ibid.

[112] P. G. Stecher, et al., *The Merck Index, Seventh Edition,* (U) (1960), ibid.

[113] P. Jeal, private communication, Atomic Weapons Establishment, Aldermaston, UK (U) (February 11, 1993).

[114] J. C. Martz, J. M. Haschke, T. J. Beugelsdjik, and L. E. Bronisz, "Automation and Integration of Site-Return Processing," in *Nuclear Materials Technology: Weapons Complex 21 Reconfiguration*, Los Alamos National Laboratory document LALP-92-41 (U) (June 1992) p. 18.

[115] F. Berkhout, A. Diakov, H. Feiveson, H. Hunt, E. Lyman, M. Miller, and F. von Hippel, *Science & Global Sec.* **3** (U) (1993) 161.

[116] A. A. Dye, private communication, EG&G Rocky Flats Plant, Golden, Colorado (U) (June 1990).

[117] D. R. Harbur, J. W. Romero, and J. W. Anderson, "Plutonium-Cerium-Cobalt Fuel Development for Molten Reactors," *AFS Meetings: Modern Castings*, **48** (U) (1965) p. 124.

[118] D. R. Harbur, J. W. Anderson, and W. J. Maraman, "Studies on the U-Pu-Zr Alloy System for Fast Breeder Reactor Applications," Los Alamos Scientific Laboratory report LA-4512, (U) (November 1970).

[119] J. M. Haschke and J. C. Martz, "Storage of Plutonium Extracted from Weapons (U)," (1993), ibid.

[120] J. D. Moseley and R. O. Wing, "Properties of Plutonium Dioxide," (U) (1965), ibid.

[121] G. Friedlander, J. W. Kennedy, and J. M. Miller, *Nuclear Radiochemistry, 2nd ed.,* (U) (1966), ibid.

[122] J. M. Haschke and J. C. Martz, "Storage of Plutonium Extracted from Weapons (U)," (SRD) (1993), ibid.

[123] J. Richter, private communication at Los Alamos National Laboratory (SRD) (March 1993).

[124] W. J. Ogle (DOE/OMA), T. J. Hirons (LANL), J. C. Mark (NRC), W. C. Myre (SNLA), and R. W. Selden (LLNL), "A DOE Special Study Report: Reactor Materials and Nuclear Explosives (U)," DOE Report, Washington, DC, DOE/DP-0003 (CNSI) (1978).